

REMARKS

Status of the Claims

Claims 1 - 21 were previously pending and at issue. Claim 1 has been amended to specify that recited layer is a layer consisting essentially of a polyester-based elastomer. Support for this amendment can be found, for example, on page 6, lines 11-12. Claims 2-5 have been amended consistent with this amendment, and to provide more correct antecedent basis.

Claim 1 has also been amended to remove the limitation regarding the shape retainability performance test. Through error, and without deceptive intent, Applicants discovered that all embodiments of the present invention did not satisfy the shape retainability performance test as previously recited in claim 1 (see Table 2 of the present invention). Claim 27 has been added which more precisely defines the shape retainability performance test. Support for this amendment can be found, for example, in the last two columns of Table 2, and the text associated therewith. Claim 28 has also been added, support for which can be found on page 10, lines 10-17 of the application as filed. No new matter has been added by these amendments. Claims 22-26 have been canceled.

Claims 1-21 and 27-28 are pending and at issue.

Rejections Under 35 U.S.C. §§ 102 and 103

Claims 1-2 stand rejected as anticipated by or obvious over Kobayashi (U.S. Patent No. 4,125,032). The Examiner states that Kobayashi discloses a heat resistant plastic tube which comprises a polyester-based elastomer including at least one of a polyester-polyester block copolymer and a polyester-polyether. The Examiner further states that the properties recited in claim 1 are necessarily present in the invention of Kobayashi since the materials and structure are the same.

Applicants respectfully disagree with the Examiner's assertion. To advance prosecution, however, claim 1 has been amended to recite at least one layer consisting essentially of a polyester-

based elastomer. Kobayashi does not disclose or suggest a layer that consists essentially of a polyester-based elastomer. Kobayashi's polyester composition contains at least 80% ethylene terephthalate. Although a small amount of elastic resin may be added to the ethylene terephthalate (preferably around 10%), the purpose of the elastic resin is to enhance the dispersibility of the organic crystallization promoting agent.

Increased crystallization leads to less elastomeric behavior (see Attached Exhibit A, *Fundamental Principles of Polymeric Materials*, Chapter 5.4, which states: "other things being equal, the greater the ratio of crystalline to amorphous phase, the stronger, harder more rigid, and less easily deformable the polymer will be"). Accordingly, Kobayashi does not disclose or suggest a layer that consists essentially of a polyester-based elastomer. Applicants request that this rejection be withdrawn.

Claims 3-5 and 16-18 stand rejected as obvious over Kobayashi in view of JP 2000290483 (JP '483). JP '483 does not disclose or suggest a polyester-polyester block copolymer with a hard segment component and a soft segment component or a polyester-polyether block copolymer with a hard segment component and a soft segment component. Kobayashi does not teach or suggest a layer that consists essentially of a polyester-based elastomer, much less such a layer that includes the recited block copolymers. Applicants request that this rejection also be withdrawn.

Claims 6-7 and 11-12 stand rejected as obvious over Kobayashi in view of Rau (U.S. Patent No. 4,510,098). Claims 8-10, 13-15 and 19-21 stand rejected as obvious over Kobayashi in view of JP '483, further in view of Rau. The Examiner cites Rau to provide a fuel feed tube usable within an engine compartment of a motor vehicle, wherein the tube further comprises a bellows portion extending at least a portion of its length. Rau does not, however, disclose or suggest the subject matter that is not taught or suggested by both Kobayashi or JP'483. More particularly Rau also does not disclose or suggest, *inter alia*, "at least one layer consisting essentially of a polyester-based elastomer including at least one of a polyester-polyester block copolymer with a hard segment component and a soft segment component and a polyester-polyether block copolymer with a hard

segment component and a soft segment component". Applicants request that this rejection be withdrawn.

In view of the above amendment, applicant believes the pending application is in condition for allowance. If an interview would be helpful to pass this application to issuance, the Examiner is requested to contact the undersigned at the number listed below.

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Respectfully submitted,

By 

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the relative amounts of the phases can vary, and this can strongly influence the bulk properties, particularly the mechanical properties. This is illustrated below for polyethylene.

Since the polymer chains are packed together more efficiently and tightly in the crystalline areas than in the amorphous, the crystallites will have a higher density. Thus, low-density (0.92 g/cm^3) polyethylene is estimated to be about 43% crystalline, while high-density (0.97 g/cm^3) polyethylene is about 76% crystalline. Density is, in fact, a convenient measure of the degree of crystallinity. Because the volumes of the crystalline and amorphous phases are additive, density and degree of crystallinity are related by

$$\frac{1}{\rho} = \frac{w_c}{\rho_c} + \frac{w_a}{\rho_a} = \frac{w_c}{\rho_c} + \frac{(1 - w_c)}{\rho_a} \quad (5.1)$$

where the w 's are weight fractions and the subscripts c and a refer to the crystalline and amorphous phases, respectively.

In the case of polyethylene, the differences in degree of crystallinity arise largely from branching that occurs during polymerization (although it is also influenced by molecular weight and the rate of cooling). The branch points sterically hinder packing into a crystal lattice in their immediate vicinity, and thus lower the degree of crystallinity.

Low-density polyethylene (LDPE) has traditionally been made by a high-pressure process (25 000–50 000 psi) and high-density polyethylene (HDPE) by a low-pressure process (~ 100 psi). Thus, LDPE is sometimes referred to as high-pressure polyethylene and HDPE as low-pressure polyethylene.

If that weren't confusing enough, low-density polyethylenes are now also made by low-pressure processes similar to those used for HDPE. These materials have most unfortunately (and inaccurately) been termed linear, low-density polyethylene (LLDPE). If they were truly linear, they wouldn't be low-density.

Traditional (high-pressure) LDPE is a homopolymer. Its long, "branched branches" arise from a side reaction during polymerization. LLDPE, however, has short branches that are introduced by random copolymerization with minor amounts (say 8–10%) of one or more α -olefins (vinyl monomers with hydrocarbon X groups) (1-butene, 1-hexene, 1-octene, and 4-methyl-1-pentene are commonly used). This same approach is extended (by adding more comonomer) to make VLDPE (very-low . . .) or ULDPE (ultra-low . . .) ($\rho < 0.915 \text{ g/cm}^3$). The nature of the branching affects some properties to a certain extent. LLDPEs form stronger, tougher films than LDPEs of equivalent density, for example. The various polyethylenes are summarized in Table 5.1 and their molecular architectures are sketched in Fig. 5.4. Equation 5.1 must be used with caution where the density is varied with a comonomer, as in LLDPE, because ρ_a will, in general, vary with copolymer composition.

Since the polymer chains are more closely packed in the crystalline areas than in the amorphous, there are more of them available per unit area to support a stress. Also, since they are in close and regular contact over relatively long

Table 5.1 The Influence of Crystallinity on Some of the Properties of Polyethylene^a

Commercial product	Low density	Medium density	High density
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Table 5.1 The Influence of Crystallinity on Some of the Properties of Polyethylene^a

Commercial product	Low density	Medium density	High density
Density range, g/cm ³	0.910–0.925	0.926–0.940	0.941–0.965
Approximate % crystallinity	42–53	54–63	64–80
Branching, equivalent CH ₃ groups/1000 carbon atoms	15–30	5–15	1–5
Crystalline melting point, °C	110–120	120–130	130–136
Hardness, Shore D	41–46	50–60	60–70
Tensile modulus, psi (N/m ²)	0.14–0.38 × 10 ⁵ (0.97–2.6 × 10 ⁸)	0.25–0.55 × 10 ⁵ (1.7–3.8 × 10 ⁸)	0.6–1.8 × 10 ⁵ (4.1–12.4 × 10 ⁸)
Tensile strength, psi (N/m ²)	600–2300 (0.41–1.6 × 10 ⁷)	1200–3500 (0.83–2.4 × 10 ⁷)	3100–5500 (2.1–3.8 × 10 ⁷)
Flexural modulus, psi (N/m ²)	0.08–0.6 × 10 ⁵ (0.34–4.1 × 10 ⁸)	0.6–1.15 × 10 ⁵ (4.1–7.9 × 10 ⁸)	1.0–2.6 × 10 ⁵ (6.9–18 × 10 ⁸)

^a It must be kept in mind that mechanical properties are influenced by factors other than the degree of crystallinity (molecular weight, in particular).

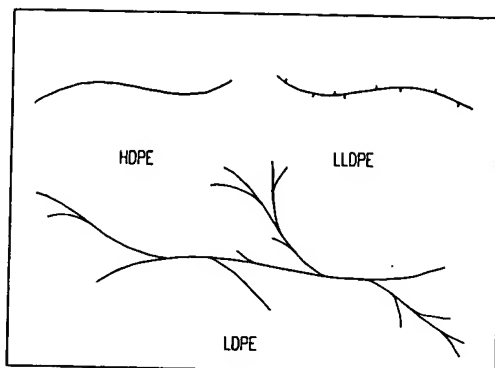


Figure 5.4 Molecular architecture of various polyethylenes. LDPE, low-density polyethylene; LLDPE, linear, low-density polyethylene; HDPE, high-density polyethylene.

distances in the crystallites, the secondary forces holding them together are cumulatively greater than in the amorphous regions. Thus, crystallinity can significantly increase the strength and rigidity of a polymer. For this reason, the stereoregular polypropylenes, which can and do crystallize, are fairly hard, rigid plastics, while the irregular, atactic polymer is amorphous, soft, and sticky. And, other things being equal, the greater the ratio of crystalline to amorphous phase, the stronger, harder, more rigid, and less easily deformable the polymer will be, as is illustrated in Table 5.1.

5.5 THE EFFECT OF CRYSTALLINITY ON OPTICAL PROPERTIES

The optical properties of polymers are also influenced by crystallinity. When light passes between two phases with different refractive indices, some of it is scattered at the interface if the dimensions of the discontinuities are comparable to or greater than the wavelength of visible light ($0.4\text{--}0.7\ \mu\text{m}$). Thus, a block of ice is transparent, but snow appears white because light must pass alternately from air to ice crystals many times. Similarly, in a crystalline polymer, the usually denser crystalline areas have a higher refractive index than the amorphous areas, so crystalline polymers are either opaque or translucent because light is scattered as it passes from one phase to the other. So, in general, *transparent polymers are completely amorphous*. An interesting exception arises in the case of isotactic poly(4-methyl-1-pentene). The refractive indices (and densities) of the amorphous and crystalline phases are almost identical. Thus, as far as light is concerned, it's a homogeneous material, and is the one transparent polymer that is known to be highly crystalline. Also, as the dimensions of the dispersed-phase particles become smaller than the wavelength of visible light, scattering decreases, so a polymer with very small crystallites and a low degree of crystallinity might appear nearly transparent, particularly in thin sections.

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The converse is not true; a polymer can be transparent due to crystallinity, but not if it contains a filler. If the polymer contains a filler, its opacity is a sure sign of crystallinity. The presence of a filler is often called "crystal" phase. Polyethylene has been synthesized in translucent packaging supports) is translucent and gas bubbles. 1–10 μm polybutadiene-styrene. Thus, while they are transparent, they have a milky appearance and appears white and

Example 3. Explain

a. Polyethylene and polypropylene are each fairly rigid, transparent, and produced in exactly the same way.

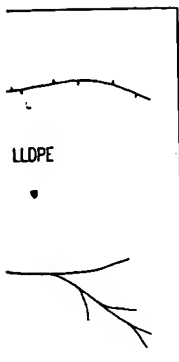
b. A plastic is similar to polyethylene and polypropylene and 35% propylene is separated by any physical means.

Solution. a. The polypropylene is thus highly crystalline. The crystallinity of the copolymer, ethylene-propylene, prevents the CH_3 groups from the propylene along the chain, preventing the formation of an amorphous, rubbery polymer.

b. Since the composition is bound within the chain, the CH_3 groups from the propylene copolymer in (a). Thus, the amorphous and stereoregular polypropylene of ethylene pack into a polypropylene lattice.

5.6 EXTENDED-CHAIN CRYSTALLINITY

Polymers crystallized from the melt have, in general, a random arrangement of chains. While the chains in an



polyethylenes. LDPE, low-density polyethylene; HDPE, high-density polyethylene.

forces holding them together are in regions. Thus, crystallinity can vary of a polymer. For this reason, the regions that do crystallize, are fairly hard, rigid; amorphous, soft, and sticky. And, in the transition from crystalline to amorphous phase, the polymer will be, gradually deformable.

OPTICAL PROPERTIES

is influenced by crystallinity. When the refractive indices, some of it is due to the discontinuities are comparable to the wavelength of light ($0.4\text{--}0.7\ \mu\text{m}$). Thus, a block of polymer is opaque or translucent because light must pass alternately through crystalline and amorphous regions. In a crystalline polymer, the refractive index is higher than the amorphous phase. So, in general, transparent polymers are amorphous. An interesting exception arises in the case of polymers with large differences in refractive indices (and densities) of the crystalline and amorphous phases. Thus, as far as light is concerned, polyethylene is the one transparent polymer that has the dimensions of the dispersed-phase comparable to the wavelength of visible light, scattering light and giving it a milky appearance. While the chains in any crystallite are oriented in a particular direction, the overall material is transparent.

The converse is not necessarily true; lack of transparency in a polymer may be due to crystallinity, but it can also be caused by an added second phase, such as a filler. If the polymer is known to be a pure homopolymer, however, translucency is a sure sign of crystallinity. Thus, commercial homopolystyrene, which is atactic and therefore completely amorphous because of the irregular arrangement of the bulky phenyl side groups, is perfectly transparent. Ironically, it is often called "crystal" polystyrene because of its crystal clarity. Isotactic polystyrene has been synthesized in the laboratory and does crystallize. It has the white, translucent appearance typical of polyethylene. Foamed polystyrene (cups, packaging supports) is white because light must pass between atactic polystyrene and gas bubbles. High-impact polystyrene consists of a dispersion of $1\text{--}10\ \mu\text{m}$ polybutadiene rubber particles in a continuous phase of atactic polystyrene. Thus, while each phase is completely amorphous and individually transparent, they have different refractive indices, so the composite scatters light and appears white and translucent.

Example 3. Explain the following facts:

a. Polyethylene and polypropylene produced with stereospecific catalysts are each fairly rigid, translucent plastics, while a 65–35 copolymer of the two, produced in exactly the same manner, is a soft, transparent rubber.

b. A plastic is similar in appearance and mechanical properties to the polyethylene and polypropylene described in (*a*), but it consists of 65% ethylene and 35% propylene units. The two components of this plastic cannot be separated by any physical or chemical means without degrading the polymer.

Solution. *a.* The polyethylene produced with these catalysts is linear and thus highly crystalline. The polypropylene is isotactic and also highly crystalline. The crystallinity confers mechanical strength and translucency. The 65–35 copolymer, ethylene–propylene rubber (EPR) is a *random* copolymer, so the CH_3 groups from the propylene monomer are arranged at irregular intervals along the chain, preventing packing in a regular crystal lattice, giving an amorphous, rubbery polymer.

b. Since the components cannot be separated, they must be chemically bound within the chains. The properties indicate a crystalline polymer, so the CH_3 groups from the propylene cannot be spaced irregularly, as in the random copolymer in (*a*). Thus, these materials must be *block* copolymers of ethylene and stereoregular polypropylene, poly(ethylene-*b*-propylene). The long blocks of ethylene pack into a polyethylene lattice and the propylene blocks into a polypropylene lattice.

5.6 EXTENDED-CHAIN CRYSTALS

Polymers crystallized from a quiescent melt (such as those in Table 5.1) will have, in general, a random orientation of the chains at the macroscopic level. While the chains in any crystallite are oriented in a particular direction,

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